

## **Understanding the Nature of Marine Aerosols and Their Effects in the Coupled Ocean-Atmosphere System**

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### **LONG-TERM GOALS**

The long-term goal of this work is to understand the nature of marine aerosol particles and how they influence visibility, cloud microphysics and precipitation, the thermodynamic structure of the marine boundary layer, and the transmission of radiation.

### **OBJECTIVES**

The objectives of this project extend across three areas: (i) advancing aerosol measurement techniques via the development of a new instrument to quantify aerosol-water interactions, (ii) improving knowledge and model predictions related to the physicochemical nature of aerosol particles and ocean-aerosol-cloud-precipitation-radiation interactions, and (iii) strengthening a research methodology leveraging multiple complementary tools of analysis to guide future studies of this nature in the marine atmosphere.

### **APPROACH**

The main technical approach is to use a combination of in-situ aircraft measurements, cloud models, and satellite remote sensors to study the nature and character of aerosols and their effects in the marine atmosphere over a broad range of spatial and temporal scales. This work includes the following tasks:

- Develop and characterize a state-of-the-art instrument to quantify aerosol-water interactions, which will be deployed in future aircraft and ship-based studies.
- Use satellite remote sensing observations to quantify and interrelate measurements of ocean bio-optical properties, aerosol and cloud microphysics, radiative properties, and precipitation in the marine atmosphere, while accounting for meteorology.
- Use cloud models to examine the salient features of cloud drop activation and the subsequent growth of drops to precipitation-sized drops over a wide range of conditions, and the capability to more easily differentiate between aerosol and meteorological effects on cloud microphysics, precipitation, and radiative transfer.

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- Carry out in-situ measurements in the marine atmosphere to simultaneously characterize aerosol physicochemical properties and to quantify the interactive nature between aerosols, oceans, clouds, precipitation, meteorology, and radiation.

Key scientists involved with this work include Dr. Fred Brechtel (Brechtel Manufacturing Inc.), who is collaborating with the PI on the development of a new instrument to quantify aerosol hygroscopicity. Dr. Graham Feingold (National Oceanic and Atmospheric Administration) has collaborated on using large eddy simulation to study aerosol effects in the marine atmosphere. Dr. Haflidi Jonsson at the Naval Postgraduate School is instrumental in the aircraft operations and the data management, which is critical for the aircraft data analysis tasks in this project. The PI collaborated with Drs. John Seinfeld (California Institute of Technology), Bruce Albrecht (University of Miami), and Lynn Russell (UC-San Diego) to conduct a combined ship-aircraft field study between July-August 2011 called the Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE).

## **WORK COMPLETED**

Project Task 1 (Instrument development): An aerosol hygroscopic growth measurement probe was developed and deployed on the Point Sur Ship during the 2011 E-PEACE field study over the eastern Pacific Ocean. Measurements were conducted with this instrument during a two week cruise along with a number of other instruments to characterize aerosol physicochemical properties. One manuscript is currently in preparation to describe the hygroscopicity instrument and present the field data collected with it.

Project Task 2/3 (Modeling and satellite data analysis): Modeling (i.e. cloud parcel model and large eddy simulation) and analysis of both aircraft and satellite remote sensing data have been completed to advance understanding of aerosol-cloud-precipitation interactions in the marine atmosphere. Five peer-reviewed publications have resulted from this project task thus far in the project lifetime, including two during this fiscal year.

Project Task 4 (Aircraft measurements): The PI is currently leading a major field study in August 2011 and was a co-PI of a field study in July 2011 to study aerosol-water-cloud-radiation interactions in the marine atmosphere over the eastern Pacific Ocean. The project is still on-going and it is anticipated that 32 total flights will be completed between July and August. A suite of state-of-the art instruments was deployed on the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter and the Point Sur Ship. Measurements of aerosol physicochemical properties (e.g. size, composition, optical properties, hygroscopicity) from both the ship and aircraft platforms will be combined with satellite data and modeling to improve knowledge of aerosol and cloud microphysics. A series of papers will be forthcoming from this project in the coming year

## **RESULTS**

Research during FY2011 has resulted in three peer-reviewed publications with highlights from these studies provided below. In addition, a new instrument was developed in collaboration with Brechtel Manufacturing Inc. to quantify aerosol hygroscopicity in the marine atmosphere. Figure 1 shows the PI's PhD student (Anna Wonaschütz) carrying out shipboard measurements with this instrument. A photograph is also shown that was taken of the Twin Otter from the Point Sur Ship. The ship coordinated the release of a smoke plume with the flight tracks of the CIRPAS Twin Otter such that

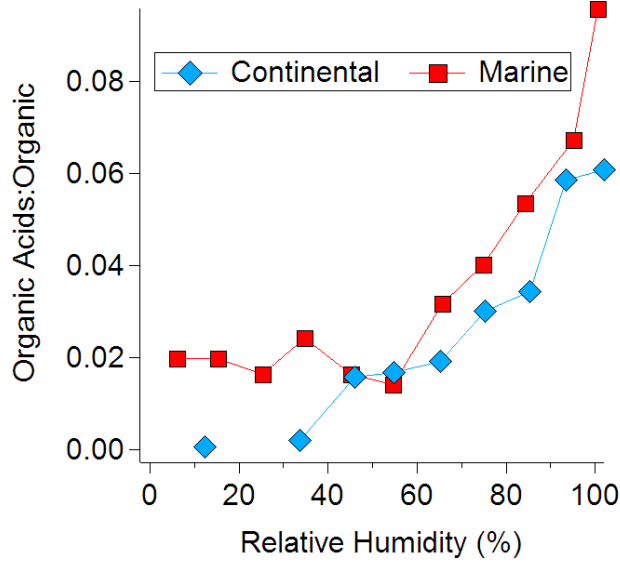
the aircraft could characterize the effects of the plume in clouds downwind of the ship. Results from this field project will be forthcoming in the next fiscal year.



***Figure 1. (Left) Doctoral student (Anna Wonaschütz) conducting shipboard measurements of aerosol hygroscopicity and chemical composition during the July 2011 Point Sur Ship cruise as part of the E-PEACE field campaign. (Right) Photograph taken of the CIRPAS Twin Otter from the Point Sur Ship, which intercepted a smoke plume emitted from the ship. One aspect of this project was to use a state-of-the-art payload of instrumentation on the Twin Otter to study the effects of the smoke plume on marine stratocumulus clouds downwind of the ship.***

*Completed Study 1: Constraining the contribution of organic acids and AMS  $m/z$  44 to the organic aerosol budget: On the importance of meteorology, aerosol hygroscopicity, and region (Sorooshian et al., 2010)*

The chemical complexity of atmospheric aerosols poses a challenge for accurate modeling of their interactions with water vapor, radiation, and clouds in the marine atmosphere. It is now well established that the organic fraction of atmospheric aerosols becomes increasingly oxidized with age, leading to species that are both less volatile and more hygroscopic. Water-soluble organic species, especially organic acids, are of interest owing to their hygroscopic properties. Airborne measurements in regions of varying meteorology and pollution are used to quantify the contribution of organic acids and a mass spectral marker for oxygenated aerosols,  $m/z$  44, to the total organic aerosol budget. Mass concentrations associated with total non-refractory organic mass and the  $m/z$  44 marker were carried out by an Aerodyne Aerosol Mass Spectrometer (AMS), while measurements of organic acids were conducted with a particle-into-liquid sampler coupled to ion chromatography (Sorooshian et al., 2006). Organic acids and  $m/z$  44 separately are shown to exhibit their highest organic mass fractions in the vicinity of clouds. The contribution of such oxygenated species is shown to increase as a function of relative humidity (Figure 2), aerosol hygroscopicity (and decreasing organic mass fraction), and is typically greater off the California coast versus continental atmospheres. Reasons include more efficient chemistry and partitioning of organic acid precursors with increasing water in the reaction medium, and high aqueous-phase processing times in boundary layers with higher cloud volume fractions. These results highlight the importance of secondary organic aerosol formation in both wet aerosols and cloud droplets.



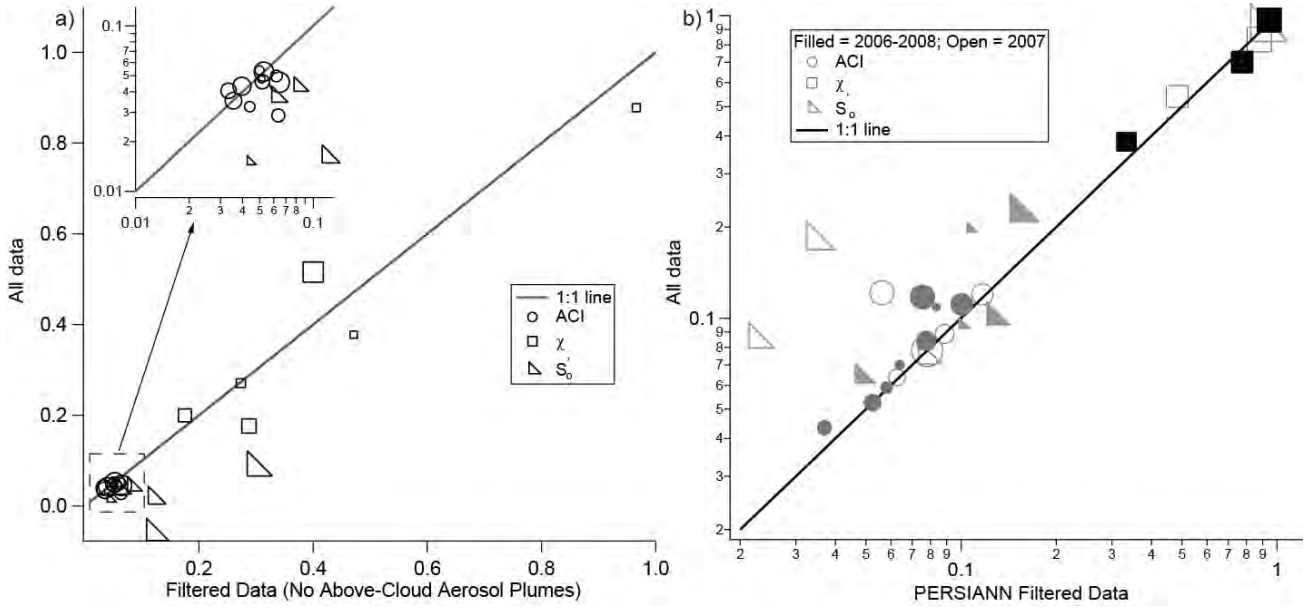
**Figure 2.** The dependence of the ratio of “organic acid:organic” mass on ambient relative humidity. The measurements were conducted using the CIRPAS Twin Otter over the eastern Pacific Ocean (“Marine”) and over southeastern Texas (“Continental”). These are the first airborne measurements showing how the organic acid mass fraction of aerosol depends on ambient relative humidity across a broad range extending from 10%-102% in the marine atmosphere.

*Completed Study 2: Investigating potential biases in observed and modeled metrics of aerosol-cloud-precipitation interactions (Duong, Sorooshian, and Feingold, 2011)*

This study utilized large eddy simulation, aircraft measurements, and satellite observations to identify factors that bias the absolute magnitude of metrics of aerosol-cloud-precipitation interactions for warm clouds. The metrics considered are precipitation susceptibility ( $S_o$ ), which examines rain rate sensitivity to changes in drop number, and a cloud-precipitation metric,  $\chi$ , which relates changes in rain rate to those in drop size. While wide ranges in rain rate exist at fixed cloud drop concentration for different cloud liquid water amounts,  $\chi$  and  $S_o$  are shown to be relatively insensitive to the growth phase of the cloud for large datasets that include data representing the full spectrum of cloud lifetime. Spatial resolution of measurements is shown to influence the liquid water path-dependent behavior of  $S_o$  and  $\chi$ . Other factors of importance are the choice of the minimum rain rate threshold, and how to quantify rain rate, drop size, and the cloud condensation nucleus proxy. Finally, low biases in retrieved aerosol amounts owing to wet scavenging and high biases associated with above-cloud aerosol layers should be accounted for and this point is emphasized in Figure 3 below. The inclusion of cases characterized by above-cloud aerosol plumes, as examined in a case study off the coast of western

Africa, is shown to depress values of  $S'_o$  ( $= -\frac{d \ln R}{d \ln \alpha}$ ) and ACI ( $= -\frac{\partial \ln r_e}{\partial \ln \alpha}$ ), where  $r_e$  is drop effective radius,  $\alpha$  is aerosol index,  $R$  is rain rate, and the partial derivatives are evaluated at fixed cloud liquid water path (LWP). On the other hand, accounting for low biases in retrieved aerosol amounts as

a result of wet scavenging with the use of an artificial neural network algorithm (PERSIANN; Sorooshian et al., 2000) is shown in some cases to result in lower values of ACI and  $S'_o$ .



**Figure 3. Satellite data analysis of the sensitivity of aerosol-cloud-rain metrics to above-cloud aerosol plumes and wet scavenging. (a) Comparison of ACI,  $\chi$  ( $= \frac{\partial \ln R}{\partial \ln r_e}$ ), and  $S'_o$  with and without above-cloud aerosol plumes off the coast of western Africa. CALIPSO satellite data were used to identify these plumes above cloud decks. The data examined range between June and October 2006. Marker sizes are proportional to LWP [11 LWP bins with up to 10% spacing around bin midpoints ( $LWP \pm 10\% \times LWP$ ), which increase in  $25 \text{ g m}^{-2}$  increments from  $50$  to  $300 \text{ g m}^{-2}$ ]. (b) Comparison of ACI,  $\chi$ , and  $S'_o$  with and without filtering of wet scavenging events prior to A-Train satellite overpasses within the same  $1^\circ \times 1^\circ$  pixel. The PERSIANN product was used to remove cases of precipitation for a period of time up to a day before a satellite overpass within the same  $1^\circ \times 1^\circ$  domain of MODIS aerosol retrievals. Marker sizes are proportional to LWP (12 LWP bins with up to 10% spacing around bin midpoints, which include  $50 \text{ g m}^{-2}$ ,  $100 \text{ g m}^{-2}$ , and up to  $1100 \text{ g m}^{-2}$  in  $100 \text{ g m}^{-2}$  increments). The data represent the JJA months for the three year period including 2006-2008 (filled) and 2007 only (open). Only points are reported in both panels that were statistically significant at 95% confidence (based on student's  $t$ -test) with and without the data filtering.**

### *Completed Study 3: Inverse modeling of cloud-aerosol interactions – Part 1: Detailed response surface analysis (Partridge et al., 2011)*

New methodologies are required to probe the sensitivity of parameters describing cloud droplet activation. This study presents an inverse modeling-based method for exploring cloud-aerosol interactions via response surfaces. The objective function, containing the difference between the measured and model predicted cloud droplet size distribution is studied in a two-dimensional framework, and presented for pseudo-adiabatic cloud parcel model parameters that are pair-wise selected. From this response surface analysis it is shown that the susceptibility of cloud droplet size distribution to variations in different aerosol physiochemical parameters is highly dependent on the aerosol environment and meteorological conditions. In general the cloud droplet size distribution is most susceptible to changes in the updraft velocity. A shift towards an increase in the importance of chemistry for the cloud nucleating ability of particles is shown to exist somewhere between marine average and rural continental aerosol regimes. This study uses these response surfaces to explore the feasibility of inverse modeling to determine cloud-aerosol interactions. It is shown that the “cloud-aerosol” inverse problem is particularly difficult to solve due to significant parameter interaction, presence of multiple regions of attraction, numerous local optima, and considerable parameter insensitivity. The identifiability of the model parameters will be dependent on the choice of the objective function. Sensitivity analysis is performed to investigate the location of the information content within the calibration data to confirm that our choice of objective function maximizes information retrieval from the cloud droplet size distribution. Cloud parcel models that employ a moving-centre based calculation of the cloud droplet size distribution pose additional difficulties when applying automatic search algorithms for studying cloud-aerosol interactions. To aid future studies, an increased resolution of the region of the size spectrum associated with droplet activation within cloud parcel models, or further development of fixed-sectional cloud models would be beneficial. Despite these improvements, it is demonstrated that powerful search algorithms remain necessary to efficiently explore the parameter space and successfully solve the cloud-aerosol inverse problem.

### **IMPACT/APPLICATIONS**

The new instrument developed to measure aerosol hygroscopicity will have application in subsequent field studies. The data collected with this instrument during the E-PEACE field study will be leveraged in future data analysis studies to advance understanding of interactions between aerosol and water vapor in the marine atmosphere. The results of Sorooshian et al. (2010) will serve to improve model parameterizations for accurately predicting the organic composition of aerosol in the marine atmosphere, which is needed to predict the radiative and hygroscopic properties of marine aerosol. Results from Duong et al. (2011) will help improve methodology in future studies attempting to examine aerosol-cloud-precipitation interactions in the marine atmosphere, especially with satellite and large eddy simulation data. The work performed by Partridge et al. (2011) demonstrates a new way to critically examine the key processes regulating cloud drop activation. The presented technique is also broadly applicable to studying other aspects of ocean-aerosol-cloud interactions.

### **RELATED PROJECTS**

There are no related projects at this time.

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## PUBLICATIONS

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## **HONORS/AWARDS/PRIZES**

Invitee: National Academy of Engineering's 2011 U.S. Frontiers of Engineering Symposium (2011)

Award for Excellence at the Student Interface (2010-2011; Dept. of Chemical and Environmental Engineering, University of Arizona)